

# Comment on “Superinsulator and Quantum Synchronization”

K. B. Efetov<sup>1,2</sup>, M. V. Feigel'man<sup>2</sup> and P. B. Wiegmann<sup>3,2</sup>

<sup>1</sup>*Theoretische Physik III, Ruhr-Universität Bochum, D-44801 Bochum, Germany*

<sup>2</sup>*L. D. Landau Institute for Theoretical Physics, Moscow 119334, Russia and*

<sup>3</sup>*James Frank Institute, University of Chicago, IL 60637, USA*

(Dated: July 31, 2008)

We show that the “theory” of “superinsulating” state presented by V.Vinokur et al (Nature vol. 452, p. 613, 2008) and Fistul et al (Phys. Rev. Lett. vol. 100, 086805, 2008 ) is essentially incorrect due to a sequence of errors in the theoretical analysis of the standard model of Josephson arrays which properties have been carefully studied and described in the literature. The line of calculations suggested in these articles lead to unphysical results. In particular, the calculations predict a direct current flowing through a capacitor. Moreover, this current may flow even in the absence of voltage - a sort of *supercurrent* flowing in the “superinsulating” state. We also question that the theoretical model employed in these works, even if treated correctly, is applicable to the analysis of experimental data on homogeneously disordered superconductive films.

1. Two recent articles<sup>1,2</sup> (hereafter referred to as I and II) are supposed to contain theory of “superinsulating” state with zero linear resistance first seen in InO<sub>x</sub> films near superconductor-insulator transition by Sambandamurthy *et al*<sup>3</sup> and later observed by Baturina *et al*<sup>4</sup> in thin films of TiN (unfortunately we found no reference in I to the first observation of this phenomenon<sup>3</sup>). Calculations presented in I and II are based on the same model and are almost identical to each other both in the method and results (few details where they differ will be mentioned below), which allows us to analyze them in parallel.

Although the materials studied experimentally in Refs.<sup>3,4</sup> are disordered homogeneous films, the authors of I and II suggested to model them by a regular 2D Josephson junctions (JJ) array. Moreover, they have arbitrarily chosen a special case when the self-capacitance of the islands  $C_0$  can be neglected with respect to the junction capacitance  $C$ . This case may be relevant to some special geometries of artificial networks but its relevance to the materials investigated in Ref.<sup>3,4</sup> has not been justified in the papers and is questionable (as will be discussed at the end of the present Comment).

Studying the model of the regular JJ array the authors of I, II claim that they discovered a “superinsulating” state with a huge dielectric gap (that grows with the size of the system) and argue that this state is due to a new phenomenon they called “quantum synchronization”.

In this Comment, we are not going to present our own interpretation of the interesting experimental results of Refs.<sup>3,4</sup>. Our aim here is to demonstrate that the theoretical part of the paper I, as well as the paper II, contains serious errors leading to wrong conclusions. Theoretical content of these papers consists both of calculations (p.2 of II and “Methods” part of I) and of a number of qualitative arguments.

We begin with analysis of these calculations and of the main result, Eqs. (1) in both papers, then we comment on some of these qualitative arguments. After that we present well-known (for about two decades) results on how the model studied in I, II really behaves at low

temperatures, and finally discuss an applicability of this particular JJ array model to the experimental data of Refs.<sup>1,4</sup>.

2. The main results of I, II are displayed in the form:

$$I \propto \exp \left[ - \frac{(\Delta_c - eV)^2 \exp(E_c/2T)}{\Delta_c E_c} \right] \quad (1)$$

$$I \propto \exp \left[ - \frac{(\Delta_c - eV)^2}{2\Delta_c T} \right] \quad (2)$$

and it is stated by the authors of I that Eq. (1) goes over to Eq. (2) at  $T \gg E_c$  (in fact it does not). Here  $I$  is the current through the system,  $V$  is the voltage and  $\Delta_c$  is the “new” gap estimated by the authors as

$$\Delta_c = \begin{cases} E_c \min \{ \lambda_c, L \} / d, & \text{for 1D arrays} \\ (E_c/2) \ln ( \min \{ \lambda_c, L \} / d ), & \text{for 2D arrays} \end{cases} \quad (3)$$

and  $\lambda_c = d\sqrt{E_{c0}/E_c} \gg d$  is the screening length ( $E_c = e^2/2C$ ,  $E_{c0} = e^2/2C_0$ ),  $L$  is the sample size;  $C_0$  is the self-capacitance of each island and  $C \gg C_0$  is the capacitance of each junction.

The “superinsulating” behavior claimed to be found in I consists of a very fast (double-exponential) temperature decrease of the conductance at low voltage  $eV < \Delta_c$  and low temperatures  $T < E_c$ , cf. (1). Eq. (3) describes a huge (especially for 1D case) gap  $\Delta_c$  depending on the size of the system and the authors argue that it is formed due to the phenomenon of the “quantum synchronization”. In fact, as we show below, Eqs. (1, 2) cannot be derived from the suggested model.

The Lagrangian  $L$  of the problem is defined by Eqs. (3,4) in I and by Eq. (4) in II. With a small modification, keeping only relevant terms considered in I and II, this Lagrangian can be written as

$$L = L_0 + L_L + L_R + L_{LR} \quad (4)$$

where

$$L_0 = \sum_{ij,kl} \left[ \frac{\hbar^2}{4E_c} (\dot{\chi}_{ij} - \dot{\chi}_{kl})^2 + E_J \cos(\chi_{ij} - \chi_{kl}) \right]$$

$$+ \sum_{ij} \frac{\hbar^2}{4E_{c0}} \dot{\chi}_{ij}^2 \quad (5)$$

and

$$L_L = E_J \sum_{j=1}^M \cos(eVt/\hbar + \chi_{1j}(t) + \psi(t)) \quad (6)$$

$$L_R = E_J \sum_{j=1}^M \cos(-eVt/\hbar + \chi_{Nj}(t) - \psi(t)) \quad (7)$$

$$L_{RL} = \frac{\hbar^2}{8E_c} \sum_{j=1}^M [\dot{\chi}_{1j} + \dot{\chi}_{Nj}]^2 \quad (8)$$

The phases  $\chi_{1j}$  and  $\chi_{Nj}$  are the leftmost and rightmost phases of the sample (next to the leads).

The authors of I and II write explicitly that, when deriving Eq. (2), they neglect all Josephson couplings inside the array, i.e. terms with  $E_J$  in  $L_0$ . (cf. paragraphs between Eqs. (12) and (13) in I and between Eqs.(8) and (9) in II).

However, once this approximation is made, the system becomes electrically disconnected for  $dc$  current between its left and right terminals. Neglecting the Josephson coupling  $E_J$  we see that the right and left parts of the system are connected to each other by the Coulomb interaction only (first term in Eq. (5)), which means that the only allowed current is the capacitive one,  $I(t) \propto C_{\text{eff}} dV(t)/dt$ , which is strictly zero in the  $dc$  limit. On a more formal level, the neglect of all the intrinsic Josephson couplings leads to the possibility of rotating the phase difference  $\chi_{1j}(t) - \chi_{Nj}(t)$  by an arbitrary constant without any energy cost (gauge invariance). Writing the corresponding expression for the current and integrating over static  $\chi_{1N}$  or  $\chi_{Nj}$  in the interval  $(0, 2\pi)$  one comes to the strictly zero  $dc$  current.

How could it happen that the authors of I, II got the nonzero direct current? We believe that it is due to an incorrect approximation made during the derivation of Eqs. (11) and (7) of the articles I and II, respectively. Namely, they first represent the sum of two cosine terms  $L_R + L_L$  in the equivalent form of the product (and then the same with the expression for the current):

$$L_R + L_L = 2E_J \sum_{j=1}^M \cos \left[ \frac{\chi_{1,j} + \chi_{N,j}}{2} \right] \times \cos \left[ \frac{2eVt + \psi + \chi_{1,j} - \chi_{N,j}}{2} \right] \quad (9)$$

and then they replace the *average of the product* of two nontrivial functions of time (e.g. Eq.(7) of I) by the *product of their averages*, thus coming to Eq. (11) of I (or Eq.(7) of II). We rewrite these equations for the current  $I_s(V)$  here for an easy reference

$$I_s(V) = A \text{Im} \int_0^\infty dt \exp[-\delta t + 2ieVt] K(t) \quad (10)$$

where  $A$  is a constant proportional to  $E_J^4$ , and the function  $K(t)$  equals

$$K(t) = \langle \exp i [\chi_1(t) - \chi_1(0) - \chi_N(t) + \chi_N(0)] \rangle_{L_0} \quad (11)$$

and  $t$  is real time.

This procedure of I and II could make sense under some special circumstances. Indeed, in the situation studied in Refs. [18,19] of I, there are no intrinsic array junctions and the autocorrelation function  $K(t) = 1$ . The same approximation could also be used if the Josephson couplings of the bulk were different from those near the leads and were sufficiently large (i.e. if the condition  $E_J \gg E_c$  would be fulfilled in the bulk of the array). In this case, phase fluctuations in the bulk would be strongly suppressed and this situation would not be essentially different from the one considered in Refs. [18,19]. This means that Eq. (10) can in some cases (when  $K(t)$  is slow function of time) be correct and the calculation based on the analog of the representation (I.11) is, in principle, possible.

However, an attempt of Vinokur *et al* (I) and Fistul *et al* (II) of using formulae written in [18,19] to their own model failed explicitly due to the neglect of cross-correlations between two trigonometric factors contained in Eq.7 of I *for vanishing values of the Josephson couplings in the bulk*. The function  $K(t)$  calculated in I, II changes with time extremely fast (on the scale of  $\Delta_c^{-1}$ ) and there is no justification of the "decoupling" approximation employed. The correct calculation *within their approximation of zero Josephson couplings inside the sample* would lead the authors to the evident and trivial result:  $dc$  current cannot flow through a capacitor.

Furthermore, Eqs. (1, 2) were written in I, II without specifying the form of the pre-exponential. Following the calculation of I, II one comes to the conclusion that the pre-exponential is proportional to  $E_J^4$  (see Eqs. (10, 11)). This is incorrect: expanding in the coupling terms of  $L_0$  one can see that the coefficient in front of such a term is strictly zero if  $N > 4$ . In fact, using the initial Lagrangian, Eqs. (4-8), a non-zero contribution to the current could be obtained (neglecting all inelastic processes as the authors of I and II did) in the  $N$ -th order of the expansion in a small parameter  $E_J/E_c$  only (where  $N$  is the size of the array).

One could wonder, however, if Eqs. (1,2) could still be derived, although with a different coefficient (not just proportional to  $E_J^4$ ).

We found that Eqs. (1, 2) cannot be repaired in such a way. The form of Eqs. (1) and (2) leads to the exponential suppression of the current not only at low voltage  $eV < \Delta_c$  but at  $eV > \Delta_c$  as well. This surprising phenomenon was not commented in paper II. At the same time, in the later paper I the authors mentioned that they used their result (1) for  $eV < \Delta_c$  only. However, such a limitation does not follow from any step made in their derivation of this result and the need to invoke it

on a purely verbal level indicates erroneous nature of the analysis.

Another problem occurs when the authors neglect contributions of non-zero winding numbers at  $T$  larger than  $E_c$  in Eq. (I.13). The quantization of charge is inherently related to the summation over all winding numbers. No insulating behavior can be obtained without it.

Actually, the necessity of taking into account the discreteness of the charges even for  $T > E_c$  is specific for the model with the vanishing self-capacitance  $C_0$  considered in the articles I, II. Adding the elementary charge  $2e$  to the system costs the huge energy  $\Delta_c$ , Eq. (3), and not  $E_c$ . Therefore, one *must* sum over the integer charges for all temperatures  $T < \Delta_c$  and the substitution of the sum by an integral is impossible. In terms of actual calculations it means that the integral over non-zero Fourier-components  $\chi_\omega$  *must* be complemented by the sum over nonzero integer winding numbers.

Neglecting winding numbers the authors take the Gaussian integral over the phase  $\chi$  and obtain (see Eq. (10) of II).

$$K(t) = \exp(-2\Delta_c E_c \zeta(T) t^2 - 2i\Delta_c t) \quad (12)$$

where  $\zeta(T) = T/E_c$  for  $T > E_c$ .

Note the presence of  $t^2$  term in Eq. (12). It is this term that might lead to Gaussian integrals and, eventually, to Eqs. (1, 2) for some physical quantities. However, the summation over the non-zero winding numbers  $k$  results in the vanishing of the  $t^2$  term in the exponent in Eq. (12). This is explicitly demonstrated, e.g., in Ref.<sup>5</sup>, where a granular metal consisting of normal grains was considered. In the absence of the  $t^2$  term in the exponent in the function  $K(t)$  one would not be able to get generally the combination  $(\Delta_c - eV)^2$  in the exponents in Eqs. (1, 2).

There is an exception when the correlation function  $K(t)$  can really be written in a Gaussian form for  $T < \Delta_c$ . This is the case of a one dimensional chain of JJ with the vanishing self-capacitance  $C_0$ . In this case, the correlation function  $K(t)$ , Eq. (11), factorizes into a product of correlation functions for each junction between the grains. The authors of I, II use this property and bring the expression for the correlation function  $K(t)$  to the form of Eq. (12) for arbitrary temperature with a function  $\zeta(T)$  proportional to  $T$  at  $T > E_c$ , and to  $\exp(-E_c/T)$  at  $T < E_c$ . Then, apparently calculating Gaussian integrals they come to Eqs. (1, 2), thus obtaining the double-exponential behavior at low temperatures. The latter is supposed to describe the “superinsulator”.

However, direct inspection shows that Eqs.(10, 12) do not lead to the results announced in Eqs.(1, 2) (at this moment we do not even discuss the validity of (10, 12)).

Substituting Eqs. (12) into Eq. (10) (Eqs. (14) or (15) of I into Eq. (11) of I) we obtain

$$I_s(V) = A \int_0^\infty \sin[2(V - \Delta_c)t] \exp(-\delta t) K_0(t) dt, \quad (13)$$

where  $K_0(t)$  is a real, even function of time defined via the relation  $K(t) = \exp(-iNE_c t/2) K_0(t)$ .

We see immediately from Eq. (13), that

$$I_s(V) \neq -I_s(-V) \quad (14)$$

In particular, Eq.(14) shows that the “superinsulating” state can carry a non-zero current without any voltage or magnetic field applied, i.e. the ground state maintains a “supercurrent”:

$$I_s(0) = -A \int_0^\infty \sin[(2\Delta_c t)] \exp(-\delta t) K_0(t) dt \quad (15)$$

It is not difficult to see that this supercurrent is even not small. Using Eq. (12) we obtain in the limit  $\delta \rightarrow 0$

$$I_s(0) \simeq -A/\Delta_c \quad (16)$$

The only way to obtain Eqs. (1, 2) from Eqs. (10, 12) is to use  $\text{Im}K(t)$  instead of  $K(t)$  in Eq. (10). But this is hard to justify. After such a replacement, for the two junction system, where  $K(t) = 1$  (in this case Eq. (10) is valid and gives a non-zero current consistent with previous works), one would get just zero current after the substitution  $K \rightarrow \text{Im}K(t)$ . The authors of I, II emphasize explicitly (between Eqs. (12, 13) in I and Eqs. (8, 9) in II) that Eq. (10) written with  $K(t)$  (not with  $\text{Im}K(t)$ ) allows them to recover the results for the two junction systems.

So, we see that following the calculational scheme of I, II one comes inevitably to the unphysical results, Eqs. (13, 16). In fact, this is a result of one more mistake made by the authors. It is clear that the non-zero “supercurrents” in the “superinsulator” originate from the non-zero value of  $\text{Im}K(t)$  obtained for the real time  $t$ . However, it follows from the definition of the function  $K(t)$ , Eq. (11), that its imaginary part is identically zero because the Lagrangian  $L_0$  is an even function of  $\chi$ , whereas the imaginary part of the exponential entering Eq. (11) is odd in  $\chi$ . Any type of averaging with  $L_0$  gives in this situation zero for  $\text{Im}K(t)$ .

Actually, the non-zero value of  $\text{Im}K(t)$  obtained in I, II is a consequence of an erroneous analytical continuation from the imaginary time  $\tau$  to the real one  $t$ . The standard procedure of the analytical continuation for computation of time-dependent quantities is as follows: i) to calculate the proper correlation function at Matsubara frequencies, ii) to make analytical continuation to real frequencies, and then, iii) to perform Fourier transformation to real times. Simple replacing  $\tau \rightarrow it$ , as was done in I, II, is not correct procedure.

Now we review few of “qualitative arguments” given in I and II.

1) In the last paragraph of left column of p.3 of II, as well as in various locations in I, the authors write that they obtained “global phase-synchronized state...” after they used the model containing no Josephson couplings inside the array. On the other hand, the authors are aware of

the duality relation between the charge and phase variables (cf. left column of p. 3 in paper I), which means the following statement: the existence of strong phase correlations implies weakness of charge number correlations, and vice versa. We are not able to understand the logics of applying (simultaneously) both the concepts of the charge quantization (which is the essence of the Coulomb blockade) and the “synchronization of phases”.

2) On the first page of I we see an estimate  $E_c \sim \Delta/g$ , where  $g \geq 1$  is a dimensionless intergrain conductance, i.e.  $E_c$  is smaller than the superconducting gap  $\Delta$  and thus smaller than the Josephson coupling  $E_J \sim g\Delta$ . We found no obvious arguments that would allow one to obtain this estimate in a model which explicitly assumes  $E_c \gg E_J$ , as we see in “Methods” part of I.

3) The statement in the 1st paragraph of the right column, p. 3 of II reads: “... even large... fluctuations in  $E_c$ ,  $E_{c0}$  and  $E_J$  as well as offset charges are negligible as compared to the huge magnitude of  $\Delta_c$ ”. We explain below that if any threshold voltage  $V_T = \Delta_c/e$  exists, it must scale with the length of the system, irrespective of any model details. Thus, the statement cited above contains a comparison of *intensive* (size-independent) quantities with *extensive* ones. To give an example: if such a statement could be correct, any local disorder would be irrelevant to the thermodynamics properties of any macroscopic system.

4) The first paragraph of p.4 in paper II contains a comparison between the lowest-temperature threshold voltage  $V_T$  and activation gap  $T_0$  measured in<sup>4</sup> at higher temperatures. In order to “explain” the high ratio  $eV_T/T_0$  the authors invoke an idea of “dielectric breakdown” and use for 2D system results of calculations obtained for 1D model. They did it since for their 2D model they obtained threshold voltage  $V_T = \Delta_c/e$  that scales just logarithmically with  $L$ , cf. second line of (3), which is not enough to get the very large ratio  $eV_T/T_0$ .

We note that: i) nothing in the model studied by the authors indicates the phenomenon of the “dielectric breakdown” raised by the authors, ii) there is no reason to invoke such a “concept” since the scaling of the threshold voltage with the length  $L$  is a direct consequence of any *correct* calculation leading to a nonzero  $V_T$ . The reason for that is simple: the relevant intrinsic parameter for a macroscopic system is the electric field  $\mathcal{E}$  and a critical value  $\mathcal{E}_T$  of this field determines the threshold. The value of  $\mathcal{E}_T$  does not depend on  $L$  for large  $L$ , whereas the voltage  $V_T \propto L$ . The authors of I,II failed to obtain such scaling in 2D due to several calculational mistakes analyzed above.

5) At last, we mention an explanation given in II, cf. Eq. (14), for the non-monotonic dependence of the insulating gap on magnetic field: it is based on a replacement of the disordered superconducting media by a *single (!) dc SQUID*, with its periodic dependence on magnetic flux through the SQUID loop. Remarkably, this “explanation” disappears completely from paper I, being replaced there by the statement that the magnetic field suppresses

the “superinsulating state” like it does with the superconducting state. That version sounds better in view of the experimental data presented in Fig.2, but it has no relation to the calculations based on the JJ array model. In addition, it does not give any hint on the non-monotonic  $B$ -dependence observed in<sup>1,3,4</sup> and “explained” in II.

3. Now we remind the readers some known properties of the model of I, II derived in a number of earlier papers<sup>6,7,8,9,10</sup>, cf. also a review article<sup>11</sup>:

a) The insulating regime is realized at low temperatures if  $E_J < aE_c$ , where  $a \sim 1$ . In this case the adequate description of charge transport involves rare hopping of Cooper pairs between the islands, as was explained already in Ref.<sup>6</sup>.

The number of these charge excitations is exponentially low at  $T \ll E_c$ , leading to the exponential suppression of the conductivity. The Coulomb interaction between the Cooper pairs in the grains was described in Ref.<sup>6</sup> using a general capacitance matrix  $C_{ij}$  without making any assumption about its parameters. This is why it was assumed that the energy of adding a particle into the system was of the order  $E_c$ .

b) The model with the vanishing self-capacitance,  $C_0 = 0$ , employed by the authors of I, II is special because the energy of interacting charges

$$E_{ch} = \frac{1}{2} \sum_{i,j} (C^{-1})_{ij} \hat{\rho}_i \hat{\rho}_j \quad (17)$$

where  $\hat{\rho}_i$  are charge operators for the Cooper pairs, is at large scales linear in 1D and logarithmic in 2D. This is precisely at the origin of the “big gap”  $\Delta_c$  found in I, II and displayed in the original form in Eq. (3). Note, however, a wrong coefficient in the second line for 2D: in fact, the energy of a single  $2e$  excitation is equal to  $\frac{2}{\pi} E_c \ln(L/d)$ . The logarithmic interaction of charges leads to a charge-binding transition of the Berezinsky-Kosterlitz-Thouless (BKT) type. The temperature  $T_c$  of this transition was estimated in Refs.<sup>7,9</sup> and<sup>10</sup>. Upon approaching this temperature from above, the linear resistivity diverges, cf. e.g. Eq. (9) of Ref.<sup>10</sup>.

c) At  $T < T_c$  all the charge excitations are paired and linear conductivity vanishes, in “dual analogy”<sup>8</sup> with the vanishing of the linear resistivity in 2D superconductors below BKT transition. Nonlinear  $I(V)$  transport at low voltage is due to electric-field induced unbinding of neutral “charge molecules”,

$$I(V) \propto (\mathcal{E}/\mathcal{E}_T)^\alpha \quad \alpha \propto T_c/T \quad \mathcal{E} = V/L, \quad (18)$$

again similar to  $V(I)$  characteristics in 2D superconductors below BKT transition; the threshold electric field can be estimated as  $\mathcal{E}_T \sim E_c/ed$ . The corresponding characteristic voltage  $V_T = L\mathcal{E}_T$  scales with  $L$  for trivial reason, being extensive variable (just like the total current through a superconductive 2D array scales with its width  $L_\perp$ ), without any “quantum synchronization” “discovered” in paper I.

4. We have arrived at the conclusion that the results obtained in I, II and expressed by Eqs. (1, 2) are incorrect. One may wonder if, however, some qualitative features of them might still be correct. Of a particular interest is the “superinsulation” formula represented by the low- $V$  limit of Eq.(1):

$$R \propto \exp \left[ \frac{\Delta_c}{E_c} \exp(E_c/2T) \right] \quad (19)$$

Since this formula was suggested in paper I for 1D case only, we analyze this specific case below and show that Eq.(19) is incompatible with simple physical arguments.

In 1D case  $\Delta_c \propto L$ , and thus, Eq. (19) demonstrates *both* the exponential dependence on the system size and much faster than exponential dependence on temperature. If such a dependence on  $T$  and  $L$  could indeed be obtained, it would be really unexpected. We show now that such a behavior does not exist.

The ground state energy is achieved when there are no charges in the system. Then Eq. (17) gives zero Coulomb energy  $U = 0$ . A configuration with the lowest non-zero energy is a dipole consisting of the charges  $+2e$  and  $-2e$  located at neighboring grains. In the model considered here the energy  $U(x)$  of the dipole of the length  $x$  linearly depends on  $x$  and, subjected to the external electric field  $\mathcal{E}$ , equals

$$U(x) = 2e(\mathcal{E}_c - |\mathcal{E}|)x \quad (20)$$

The energy of the dipole linearly grows with  $x$  for  $|\mathcal{E}| < \mathcal{E}_c$ . A current through the system is possible provided the size of the dipole can reach the size of the system  $L$ . The energy corresponding to such a dipole is huge:

$$U(L) = 2(\Delta_c - eV) = 2eL(\mathcal{E}_c - |\mathcal{E}|) \quad (21)$$

where  $\mathcal{E}_c = \Delta_c/eL$ . However, the probability of creating this dipole is finite at finite  $L$  and proportional to  $\exp(-U(L)/T)$ , therefore the resistivity  $R$  can be written as

$$R \propto \exp \left[ \frac{2eL(\mathcal{E}_c - |\mathcal{E}|)}{T} \right] \quad (22)$$

We see from Eq. (22) that the resistivity obtained from this simple consideration contains the combination  $\Delta_c - eV$  and not square of it. Therefore, the Coulomb blockade is important for  $eV < \Delta_c$  only.

Eq. (22) shows that the Coulomb blockade can be overcome if one creates a dipole with the energy of order  $\Delta_c$ . Clearly, this probability is small but it is described by the activation law. This simple argument excludes the double-exponential temperature dependence of the resistivity  $R$ , Eq. (19).

We want to emphasize that the big gap in the spectrum of the excitations  $\Delta_c$  exists (within the specific 1D model discussed) not only for the Cooper pairs but for the normal electrons as well<sup>5</sup> (in the latter case one would

have to use  $\Delta_c/4$  instead of  $\Delta_c$ ). So, if the “superinsulation” was due to an arbitrarily chosen special form of the capacitance matrix, it would exist even without any superconducting pairing.

An important remark is in order: following I and II we considered the situation of an *exponential* dependence of resistivity on the system size  $L$ . In reality, this usually does not make sense for large systems: in 2D case a resistance *per square*  $\rho(T)$  can be defined, whereas in 1D case the resistance grows  $\propto L$ . This behavior is due to inelastic processes including thermal activation. Such processes (neglected within the calculational scheme used in papers I and II) lead, in particular, to the current-voltage dependence (18) for 2D case. 1D model with weak  $E_J$  and vanishing self-capacitance is very specific due to its simplicity: it reduces to a single capacitor with  $C_{\text{eff}} = C/N$ . Thus, the meaning of Eq. (22) is also trivial: it corresponds to a resistance through a capacitor in the Coulomb blockade regime. Actually, non-trivial macroscopic systems with a low-temperature conductivity proportional to  $\exp(-cL)$  do exist: such a behavior is a consequence of a “quantum topological order”, cf. e.g.<sup>12</sup>. The standard 2D Josephson-junction model discussed in I, II does not lead to such a behavior.

5. Could this 2D regular JJ array model be used to describe experimental data<sup>3,4</sup> on homogeneously disordered superconductive films? The authors of I wrote (top of the last column of the main text): “our understanding of the origin of superinsulating state in the films relies on the formation of the network of superconducting droplets within the normal matrix”. Still, the model they considered contains superconductive islands connected by tunnel barriers without any sign of normal matrix inside. The problem of the superconductive islands put into a normal matrix is essentially different in its physics and more complicated for theoretical studies<sup>13,14,15,16</sup> in comparison to the model of 2D JJ array; its relevance to the data of<sup>3,4</sup> could be a subject of separate work. Coming back to the discussed model of 2D JJ array, we note its serious deficiencies regarding a possibility of an application to homogeneously disordered superconductors:

- i) in the absence of well-defined (structural) grains separated by tunnel junctions, the notion of the charging energy  $E_c$  becomes ill-defined, to begin with;
- ii) even if the grains separated by tunnel barriers are assumed to appear “somehow” in a continuously disordered media, there is no reason to expect them to form a regular lattice with identical areas,  $E_J$  and  $E_c$ .
- iii) even if in spite i) and ii) a granular model could with some degree of imagination be thought of as relevant, the model considered in I, II contains a vanishing ratio of the capacitances  $C_0/C$  and this is the crucial point to obtain the “huge” gap; why should this condition be fulfilled in a “self-organized” granular network?
- iv) care should be exercised with respect to the role of random stray charges, which are always present in real arrays and influence strongly the insulating part of the phase diagram for any model where the insulating be-

havior is due to Coulomb repulsion (cf. e.g.<sup>17,18</sup> and references therein). In particular, in the presence of a charge disorder with a typical amplitude of order  $e$ , the charge-pairing BKT-like transition does not exist.

Based on these arguments we question any kind of “fitting” the experimental data<sup>4</sup> by results of *any* calculations based on the simplified JJ array model.

To summarize: the phenomenon dubbed as “quantum synchronization” does not exist in the model studied. This model describes the well known phenomena of Coulomb blockade. The result obtained by the authors are in conflict with previous studies of the model and we believe are based on erroneous calculations.

- 
- <sup>1</sup> V. M. Vinokur, T. I. Baturina, M. V. Fistul, A. Yu. Mironov, M.R. Baklanov, and C. Strunk, *Nature*, **452**, 613 (2008)
  - <sup>2</sup> M. V. Fistul, V. M. Vinokur and T. I. Baturina, *Phys. Rev. Lett.* **100**, 086805 (2008).
  - <sup>3</sup> G. Sambandamurthy, L.W. Engel, A. Johansson, E. Peled and D. Shahar, *Phys. Rev. Lett.* **94**, 017003 (2005).
  - <sup>4</sup> T. I. Baturina, A. Mironov, V. M. Vinokur, M.R. Baklanov, and C. Strunk, *Phys. Rev. Lett.* **99**, 257003 (2007).
  - <sup>5</sup> K.B. Efetov and A. Tschersich, *Phys. Rev. B* **67**, 174205 (2003)
  - <sup>6</sup> K.B. Efetov, *Zh. Eksp. Teor. Fiz.* **78**, 2017 (1980) (*Sov. Phys. JETP* **51**, 1015 (1980))
  - <sup>7</sup> J. E. Mooij *et al*, *Phys. Rev. Lett.* **65**, 645 (1990)
  - <sup>8</sup> M. P. A. Fisher, *Phys. Rev. Lett.* **65**, 923 (1990).
  - <sup>9</sup> R. Fazio and G. Schoen, *Phys. Rev. B* **43** 5307 (1991)
  - <sup>10</sup> M. V. Feigel'man, S. E. Korshunov and A. B. Pugachev *Pisma v ZhETF*, **65**, 541 (1997)[*JETP Lett.* **65**, 566 (1997)].
  - <sup>11</sup> R. Fazio and H. van der Zant, *Phys. Rep.* **355**, 235 (2001).
  - <sup>12</sup> L.B.Ioffe and M.V.Feigel'man, *Phys. Rev. B* **66**, 224503 (2002).
  - <sup>13</sup> M. V. Feigelman and A. I. Larkin, *Chem. Phys.* **235**, 107 (1998)
  - <sup>14</sup> B. Spivak, A. Zyuzin and M. Hruska, *Phys. Rev. B* **64**, 132502 (2001)
  - <sup>15</sup> M. V. Feigelman, A. I. Larkin and M. A. Skvortsov, *Phys. Rev. Lett.* **86**, 1869 (2001).
  - <sup>16</sup> M. A. Skvortsov and M. V. Feigelman, *Phys. Rev. Lett.* **95**, 057002 (2005)
  - <sup>17</sup> E. Granato and J. M. Kosterlitz, *Phys. Rev. B* **33**, 6533 (1986).
  - <sup>18</sup> I. V. Protopopov and M. V. Feigelman, *Pisma v ZhETF* **85**, 621 (2007) [*JETP Lett.* **85**, 513 (2007)].

## I. NOTE ADDED ON JULY 30, 2008.

Recently, M. V. Fistul, V. M. Vinokur, T. I. Baturina (FVB) submitted a more detailed paper III (*Macroscopic Coulomb blockade in large Josephson junction arrays*, arXiv:0806.4311, v1). Several formulae criticized in our comment have been corrected. In III the current  $I_s(V)$  is expressed through a retarded correlation function of hopping amplitudes and it is shown how to write this function at real time. This makes the current an odd function of the voltage  $V$  as it should be.

However, the most serious errors remain. A major approximation when the first cosine in Eq. (9) (of our Com-

ment) is replaced by a constant would be justified if the Josephson couplings in the bulk were large. However, the authors consider an opposite limit. There the phase difference between grains in the bulk strongly fluctuates with time and the stationary perturbation theory used in III, Eqs. (10-13), is not applicable. For the same reason, the arguments lead to Eq. (31) of III are not consistent. Setting all couplings in the bulk to zero, as the authors do, breaks contacts between grains. This system (thus approximated by a capacitor) cannot support a finite  $dc$  current.

Nevertheless, in papers I-III the authors present formulae for  $dc$  current as a smooth function of voltage  $V$  (see Eqs. (46, 54) of III). How could it happen?

We have found that an integral obtained as a result of their approximation has not been calculated properly.

Following I-III one can write the current in the form of Eq. (25) of III with the correlation function  $K(t)$  written in the first equation of Eqs. (33) of III (or, Eq. (52) of III). For a large number of grains one can neglect an influence of the leads and put  $K_{leads}(t) = 1$ . The integral obtained in this way can be easily evaluated in different limits.

For example, in the limit  $E_c/\ln N \ll T \ll E_c$ , where  $N$  is the number of the grains in the system, the authors of III obtained their Eq. (53). They calculate the integral using the saddle point method: expanding  $\cos(E_c t)$  entering Eq. (53) in  $t$  near  $t = 0$  and then computing the Gaussian integral around this saddle point. In this way the authors obtained the double exponential behavior (see Eq. (1) of our comment).

However,  $t = 0$  is not the *only* saddle point of this integral: all  $t = \pm 2\pi m$  with integer  $m$  are saddle points as well, and all of them must be taken into account. Integrating near all the saddle points and summing them up yields the following expression for the current

$$I_s(V) = \frac{\pi^{3/2} B}{\sqrt{N}} e^{E_c/4T} (I_0(V) - I_0(-V)), \quad (23)$$

where

$$I_0(V) = \sum_{k=-\infty}^{\infty} \delta(2eV - E_c(N/2 + k)) \exp\left(-\frac{k^2}{4N} e^{E_c/2T}\right)$$

and  $B$  is a coefficient proportional to  $E_J^4$ .

Eq. (23) contains a sum of matrix elements between the states with different numbers of the particles of *iso-*

*lated* grains. The discreteness of the levels is the consequence of the discreteness of the charge of the Cooper pairs. It is there at all temperature regimes once contacts are broken. The levels may be smeared only by taking into account charge tunnelling between the grains which has been explicitly ignored by the approximation made by the authors. A tunnelling (a non-zero  $E_J$ ) smoothes  $I(V)$  functions, not the temperature. This, seems to be an obvious point, has been discussed in the main part of our Comment (see e.g., Eqs. (18,22)). No disorder can help in this respect as long as the contacts remain broken.

Correct calculation of the integral does not save Eq. (23), though. It still shows a dc-current flowing through

a capacitor at resonant values of voltage.

Finally we comment on the last paragraph on p.11 of III. There the authors explain how one should think of “superinsulating” state: the internal part of the array (except the rightmost and leftmost junctions) “acts coherently as a single superconducting island”. It is not quite clear whether the authors have in mind a closed ring to be a superconductor or “superinsulator”. Regardless of this, we do not think that superconductivity in an array with broken junctions is possible anyway.

We conclude that the new theoretical material presented in III neither clarifies nor essentially corrects the analysis and the results of I and II.